

Shipper/Receiver Difference Verification of Spent Fuel by use of PDET

Y. S. Ham, S. Sitaraman

June 6, 2011

The Second International Conference on Advancements in Nuclear Instrumentation, Measurement Methods and their Application

Ghent, Belgium

June 9, 2011 through June 10, 2011

Disclaimer

This document was prepared as an account of work sponsored by an agency of the United States government. Neither the United States government nor Lawrence Livermore National Security, LLC, nor any of their employees makes any warranty, expressed or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States government or Lawrence Livermore National Security, LLC. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States government or Lawrence Livermore National Security, LLC, and shall not be used for advertising or product endorsement purposes.

Shipper/Receiver Difference Verification of Spent Fuel by use of PDET

Young S. Ham and Shivakumar Sitaraman

Abstract- Spent fuel storage pools in most countries are rapidly approaching their design limits with the discharge of over 10,000 metric tons of heavy metal from global reactors. Countries like UK, France or Japan have adopted a closed fuel cycle by reprocessing spent fuel and recycling MOX fuel while many other countries opted for above ground interim dry storage for their spent fuel management strategy. Some countries like Finland and Sweden are already well on the way to setting up a conditioning plant and a deep geological repository for spent fuel. For all these situations, shipments of spent fuel are needed and the number of these shipments is expected to increase significantly. Although shipper/receiver difference (SRD) verification measurements are needed by IAEA when the recipient facility receives spent fuel, these are not being practiced to the level that IAEA has desired due to lack of a credible measurement methodology and instrument that can reliably perform these measurements to verify non-diversion of spent fuel during shipment and confirm facility operator declarations on the spent fuel.

In this paper, we describe a new safeguards method and an associated instrument, Partial Defect Tester (PDET), which can detect pin diversion from Pressurized Water Reactor (PWR) Spent Fuel Assemblies in an in-situ condition. The PDET uses multiple tiny neutron and gamma detectors in the form of a cluster and a simple, yet highly precise, gravity-driven system to obtain underwater radiation measurements inside a Pressurized Water Reactor (PWR) spent fuel assembly. The method takes advantage of the PWR fuel design which contains multiple guide tubes which can be accessed from the top. The data obtained in such a manner can provide spatial distribution of neutron and gamma flux within a spent fuel assembly. Our simulation study as well as validation measurements indicated that the ratio of the gamma signal to the thermal neutron signal at each detector location normalized to the peak ratio of all the detector locations gives a unique signature that is sensitive to missing pins. The signature is principally dependent on the geometry of the detector locations, and little sensitive to enrichment or burn-up variations. A small variation in the fuel bundle, such as a few missing pins, changes the shape of the signature to enable detection. After verification of the non-diversion of spent fuel pins, the neutron signal and gamma signal are subsequently used to verify the consistency of the operator declaration on the fuel burn-up and cooling time.

Manuscript received May 20, 2010. This work was performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under Contract DE-AC52-07NA27344, and supported by Next Generation Safeguards Initiative Program, Office of Nuclear Safeguards & Security and Radiological Source Replacement and Global Nuclear Safeguards R&D Programs, Office of Nonproliferation Research & Development, National Nuclear Security Administration, Department of Energy.

Y.S. Ham and S. Sitaraman are with Global Security Directorate, Lawrence Livermore National Laboratory, Livermore, CA 94550 USA (ham4@llnl.gov)

I. INTRODUCTION

The objective of snipper/receiver and measurements is to ensure that the nuclear material that The objective of shipper/receiver difference (SRD) leaves a shipping facility remains identical without any diversion or modification when it arrives at the receiving facility. SRD of nuclear material is typically measured at the receiving facility as soon as the nuclear material has arrived at the receiving facility. When the nuclear material is spent fuel, the SRD measurement is extremely difficult or impossible as there is no practical instrument that confirms the shipper information. In practice for the case of reprocessing plants, SRD measurements have to wait until head end processes, which include chopping, shearing of spent fuel into small pieces for dissolution in nitric acid. are completed on the spent fuel assembly and the spent fuel is transferred to the input accountability tank. SRD is defined by IAEA as the difference between the quantity of nuclear material in a batch as stated by the shipping material balance area and as measured at the receiving material balance area.

As the shipper's values are based on the calculated fissile contents of spent-fuel assemblies at the reactor, whereas receiver's values are based on measurements at the input accountability tank that are adjusted for losses in hulls, there exists uncertainty or discrepancy of the reactor operator calculations of plutonium content up to 5-10 percent. This creates a situation that SRD can be up to 5-10% even if no material has been lost or diverted. Verification of shipper's data such as contents of plutonium or uranium in a spent fuel assembly is practically impossible as the verification requires verification of fuel manufacturer's data, the knowledge of the detailed reactor power operating history as well as the shipper's isotopic generation and depletion calculation methods. Even if the entire shipper's data are trustworthy, the discrepancy between declared values and measured values has to be reconciled without compromising safeguards principles. Improvement of the measurement techniques alone at the accountability tank does not necessarily solve this fundamental problem as the computer calculations have to be perfect, something that is difficult to achieve.

One manifestation of this situation is that the discrepancy of the input material in the accountancy tank at THORP of BNFL was only found after 8 months of steady loss of material through a leak in the accountancy tank, which amounted to 20 tonnes of uranium and 200 kilograms of plutonium. Interestingly the leak was found because of calculated

discrepancies in the nuclear material balance that had been performed for safeguards purposes in April, 2005 [1-2]. This accident demonstrates that diversion can go unnoticed if diversion of fuel pins from spent fuel assemblies ranging between 10 and 20% with some adjustment on the shipper's values or even potentially without any adjustment on the shipper's values if the degree of diversion is somewhere below 5%. The diversion can happen anytime during the lifetime of spent fuel even during reloading prior to their arrival at the reprocessing plant. In such a scenario, these spent fuel assemblies will be reprocessed without raising any suspicion of material diversion.

The issue of SRD is not just limited to reprocessing plants. In fact, recipient facilities that receive spent fuel are all subject to the issue of SRD determination. Examples of this would

include conditioning plants that receive spent fuel for geological repositories, pyroprocessing facilities, or dry storages. Often the spent fuels are accepted into these facilities without thorough evaluation of partial defect testing, a serious safeguards and security issue that need to be addressed.

In this paper we describe a new method that addresses the issue of determination with implementation of an instrument under development, Partial Defect Tester (PDET), which can detect pin diversion from PWR spent fuel assemblies in an in-situ or isolated condition. The PDET uses multiple tiny neutron and gamma detectors in a form of cluster and highly precise, gravity-driven system to obtain simultaneously underwater radiation measurements inside guide tubes of a Pressurized Water Reactor (PWR) fuel assembly. spent The information gathered is used to detect pin diversion as well as to confirm consistency of the facility provided data.



Fig. 1: A picture of PDET

II. METHODS

A. Partial Defect Verification by PDET

Every PWR fuel assembly has as a design feature a set of guide tubes where a control rod assembly can be inserted (see Fig. 1.). The control rod assembly is used to control neutron flux during reactor operation. In the discharged spent fuel assembly (SFA), the guide tubes are filled with water when stored in the spent fuel pool. The concept of partial defect verification is to use the gamma and neutron flux information inside these guide tube holes to develop signature profiles that are invariant in intact SFAs.

The gamma and neutron signals are obtained by inserting tiny neutron and gamma detectors into the guide tubes of a SFA. The guide tubes form a quadrant symmetric pattern in the various PWR fuel product lines and the neutron and gamma signals from these various locations are processed to obtain a unique signature for an undisturbed fuel assembly, defined as the base signature. The base signatures can be formed from gamma signals, neutron signals or gamma to neutron ratio. The base gamma signature is the arrangement of the gamma signals at each of the guide tube locations normalized to the maximum among them in a particular pattern. For example, for a 14x14 PWR SFA, there are 16 guide tubes, and thus 16 measurement positions or 16 gamma data points. A symmetric pattern or base signature is obtained when gamma signals are plotted in a systematic manner starting with the guide tube location closest to the center and moving in a counter-clockwise manner for each cluster of 4 guide tubes (e.g. c, d, a, b, etc.) Figure 2 shows the alphabetic labels 'a' through 'p' for the sixteen locations. The base signatures of neutron and ratio of gamma to neutron are obtained in a similar manner. Figure 3 shows a typical base signature for the ratio when the SFA has no missing fuel pins. In the case of diversion of nuclear fuel pins, one or more of the base signatures gets distorted and the amount of distortion depends on the degree of diversion.

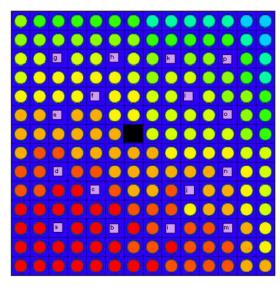


Fig. 2: Fuel lattice with guide tube location

Previous papers detailed the development of this unique signature that will be noticeably perturbed if some of the fuel pins are replaced with dummy pins both in isolated SFAs as well as SFAs in an in-situ condition in the storage racks in symmetric or random removal patterns [3-6]. The methodology was validated with measurements in SFAs with excellent agreement between the experimental and simulated data. Thus a visual inspection of the signature can identify partial defects, making the verification method easy to interpret without requiring operator declared data or fuel movement [7].

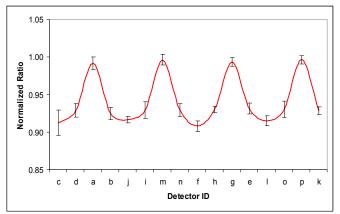


Fig. 2. Typical base ratio signature produced by the normalized gamma to thermal neutron ratio

B. Shipper/Receiver Verification Methodology

SRD verification methodology is based upon using thermal neutron and gamma dose data generated by PDET measurements. We started with a hypothesis that there is a unique relationship between burnup and neutron count similar to the relationship between burnup and neutron count generated by FDET [8] as well as potentially a unique relationship between total gamma dose and burnup.

In order to explore those relationships, a set of simulations was performed using a 14x14 Westinghouse PWR spent fuel assembly to obtain the gamma and neutron signals at the sixteen guide tube locations. These sixteen guide tubes represent locations where measurements can be made by inserting tiny gamma and neutron detectors. Each simulation in the set was performed by using uniform burnups ranging from 15-45 GWd/t, an initial enrichment of 3.8 w% ²³⁵U, and a ten year cooling time. In addition two more simulations were performed for assemblies with varying intra-assembly burnups in the range of 20-40 GWd/t. These two PWR assemblies were actual discharged PWR assemblies from a commercial nuclear power plant. The neutron and gamma source strengths for the assembly as well as isotopics at each burnup level were obtained using the burnup and decay code, ORIGEN-ARP [9]. The Monte Carlo radiation transport code, MCNP [10], was used to obtain the gamma and neutron signals at the guide tube locations.

The average gamma signal based on the sixteen locations was plotted against the burnup and linearly fitted to obtain an expression relating gamma signal and burnup. In addition, the average gamma signal of the four guide tube locations closest to the center of the assembly was also plotted against burnup.

The gamma dose relationship with burnup can be expressed as

$$G(bu) = a * (bu) + b$$

where G is gamma dose which is Cs137 cooling time corrected, bu is fuel burnup, a and b are constants. It is assumed that spent fuel assemblies are at least several years old such that gamma dose is principally Cs137 dominated.

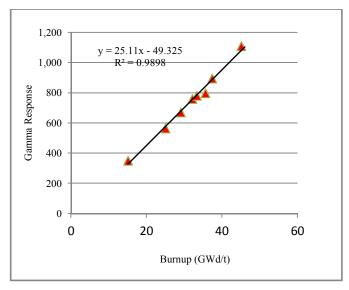


Fig. 3. Simulated gamma dose data based on average of entire 16 guide tube locations as a function of burnup.

The same linear relationship holds true when the average data at four locations were used. These four locations will be minimally impacted by gammas coming in from neighboring assemblies in an *in-situ* condition where the test assembly would be surrounded by other in the storage pool. They would thus represent a more realistic basis for confirming the operator declared burnup as well as the fact that the gamma signals at these central locations would represent the average burnup of the assembly in cases where there is an intra-assembly burnup gradient.

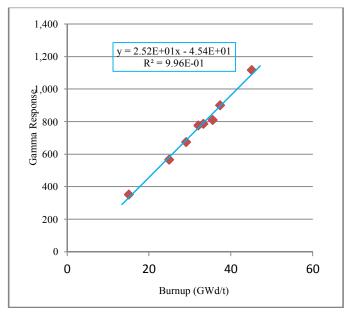


Fig. 4. Simulated gamma dose data based on average of 4 guide tube locations as a function of burnup.

Note the correlation between total gamma dose and burnup was very linear, although total gamma dose was used for gamma signal, whether the average gamma dose was used based on sixteen guide tube locations or four guide tube locations. This is an extraordinary finding that renders SRD verification very practical and *in-situ* verification possible.

Similar to gamma data generation, simulated thermal neutron data were also generated at the guide tube locations producing two curves shown in Figs 5 and 6.

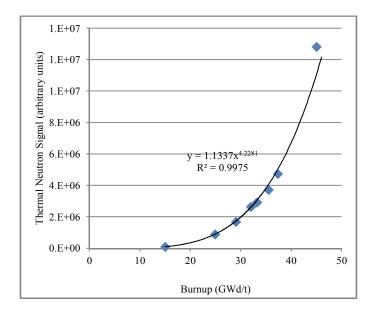


Fig. 5. Simulated thermal neutron signal based on average of entire 16 guide tube locations as a function of burnup.

The neutron signals exhibited a strong power function of burnup (~4.3) similar to a power function generated by FDET [8]. Note that the power function for neutrons is sensitive to the initial enrichment of the fuel.

The neutron count relationship with burnup can be expressed as

$$N(bu) = c * (bu)^d$$

where N is neutron count that is Cm244 cooling time corrected, bu is fuel burnup, c and d are constants.

Having established two relationships or "calibration curves" between burnup and gamma dose, and between burnup and neutron data, the curves can be used to confirm facility operator declared cooling time and burnup values. An IAEA inspector can easily choose a data collection method depending upon facility situations. If spent fuel assemblies can be lifted and isolated for measurements, both neutron and gamma data are used for partial defect testing, and then confirmation for cooling time and burnup information using both "calibration curves. It is impossible for an operator to satisfy both calibration curves in the case of trying to cheat by changing both cooling time and burnup data. Isolated measurements of spent fuel are the only way to ensure nondiversion of spent fuel pins and to confirm both cooling time and burnup. If only in-situ measurements are possible, partial defect testing is still performed first, but cooling time and burnup information are confirmed only by the calibration curve that uses gamma information. However, there is still a potential to satisfy the calibration curve by simultaneously changing burnup and cooling time data.

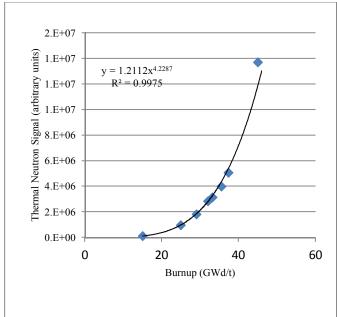


Fig. 6. Simulated thermal neutron signal based on average of 4 guide tube locations as a function of burnup.

III. EXPERIMENTS

In order to validate the concept of the SRD verification methodology, experiments were carried out to obtain gamma signals inside guide tubes of PWR spent fuel assemblies which were being stored at a commercial spent fuel pool at a reactor site. Note that measurement data were obtained *in-situ* without moving any spent fuel assemblies individually at each guide tube position at the depth of 1.25 meter below the top nozzle. Table 1 shows the fuel information of the of the six Westinghouse type PWR fuel assemblies used for measurements.

In-house developed underwater neutron measurement system was used to measure neutron signals inside guide tube holes in PWR spent fuel assemblies. A Centronic fission chamber was used for thermal neutron measurements in a waterproof housing, and IAEA standard electronics and software, i.e., Mini Multi-Channel Analyzer and WinMCS software, for data acquisition. For gamma measurements, a special type of ion chamber was fabricated by Technical Associates. The ion chamber was waterproof and could be directly inserted into guide tubes. Whereas a computer and data acquisition software were needed for thermal neutron measurements, the gamma radiation dose could be directly read from a dose reader in a digital format. Typically it took less than a few minutes for insertion of neutron detector, and additional 900 seconds for a single position measurement in MCS mode.

Table I: Description of the six 17x17 PWR spent fuel assemblies used for experiments.

Fuel ID	Burnup (GWd/tU)	Discharge Date	Initial Enr't (%)
A01	13.373	7/30/86	1.61
B50	21.611	11/12/87	2.41
Y02	31.949	1/16/92	3.09
G29	38.957	1/2/93	3.23
H26	41.892	3/20/94	3.49
H13	42.138	3/20/94	3.5

IV. RESULTS AND DISCUSSION

Table II shows the measurement data at four guide tube locations of the spent fuel assemblies, average gamma dose, and Cs137 corrected gamma dose. Variation in the four data of an assembly indicates the degree of burn-ups within an assembly.

Measured gamma dose data and Cs137cooling time corrected data are plotted in Fig. 7 as a function of burnup. Cs137 cooling time corrected plot shows a good linearity demonstrating that confirmation of the operator declared data of burnup and cooling time can be achieved by creating and using a "calibration curve". Spent fuel assemblies with erroneous data will deviate from this "calibration curve". The reason for linearity of the raw data is due to relatively close discharge date compared to the long half life of Cs137.

In order to look at the simulated and measured data on a same plot, the simulated data were normalized to the measured data by minimizing the square error of the estimate, resulting in the normalization factor, E

$$E = \frac{\sum_{i} x_i r_i}{\sum_{i} x_i^2}$$

where x_i and r_i are the simulated and measured responses, respectively.

Figures 8 shows the simulated and measured data together with the fitted equation for the measured data for the cases of the average simulated gamma signal based on the average simulated gamma signal at the four guide tube locations closest to the center of the assembly.

One can observe that the "calibration curve", Cs137 corrected plot, is relatively insensitive to initial enrichment variation of the fuel. This feature is valuable in actual field verification in particular when only a small number of

measurements is to be performed due to limited time available for measurements at a facility.

Although neutron data are not useful in the analysis as measurements were done *in-situ*, neutron data are, in general, much more sensitive to initial fuel enrichment. For this reason, the spent fuel assemblies with the same initial enrichment need to be grouped together for their analysis.

In an actual verification with the use of PDET, an inspector would choose a method of measurements either *in-situ* or in an isolated measurement condition. Initially the measured data are used for partial defect verification of the spent fuel assembly. Subsequently, all measured data are plotted as a function of burnup as shown in Fig. 7 or Fig. 8 to confirm cooling time and burnup of every spent fuel assembly.

Table II: Measurement data at four guide tube locations within spent fuel assemblies along with average and Cs137 corrected gamma response.

Fuel ID	Gamma Dose at four locations (relative)			Measured Average Gamma	Cs137 Corrected Gamma	
A01	183	184	183	182	183	310.7
B50	268	297	324	295	296	487.9
Y02	467	486	510	486	487	729.2
G29	660	661	631	633	646	944.9
H26	703	714	737	732	722	1027
H13	715	733	722	699	717	1020

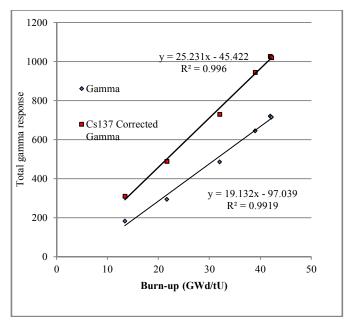


Fig. 7. Measured gamma dose data and Cs137 corrected data based on 4 guide tube positions as a function of burnup. Both plots show good linearity suggesting that the methodology can be a good way to verify declared burnup.

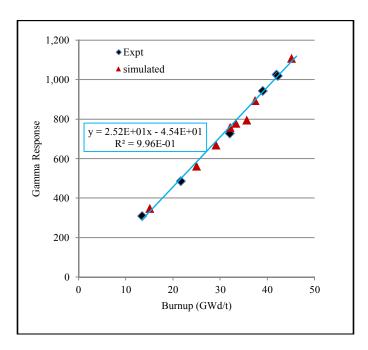


Fig. 8. Comparison of measured and simulated data based on four guide tube locations

V. CONCLUSION

An effective methodology has been developed to confirm SRD verification utilizing thermal neutron and gamma dose information collected at guide tube locations of a PWR spent fuel assembly. The data can be generated easily by PDET. The method can be applicable either in an isolated spent fuel measurement condition or *in-situ* condition. The PDET armed with this new cooling time and burnup verification method can

be a powerful and yet a practical tool to ensure integrity of spent fuel that it encounters.

REFERENCES

- Annual Report and Accounts 2005, Radiological Protection Institute of Ireland, 2005
- [2] Court transcript of a case against BNFL, 16 Oct, 2006, http://www.greenpeace.org.uk/MultimediaFiles/Live/FullReport/8065.p
 df
- [3] Young S. Ham and Shivakumar Sitaraman, "Partial Defect Tester (PDET) - A Novel Approach to Detect Partial Defects in Pressurized Water Reactor Spent Fuel", Nuclear Technology (accepted for publication)
- [4] Sitaraman, S. and Ham, Y., "Characterization of a Safeguards Verification Methodology to Detect Pin Diversion from Pressurized Water Reactor (PWR) Spent Fuel Assemblies using Monte Carlo Techniques," Proceedings of the INMM 48th Annual Meeting, Tucson, AZ, July 2007.
- [5] S. Sitaraman and Y.S. Ham, "Sensitivity Studies for an In-Situ Partial Defect Detector (PDET) in Spent Fuel using Monte Carlo Techniques", 49th Annual Meeting of the Institute of Nuclear Materials Management, Nashville, TN, July 2008.
- [6] S. Sitaraman and Y.S. Ham, "Symmetric Pin Diversion Detection using a Partial Defect Detector (PDET)," 50th Annual Meeting of the Institute of Nuclear Materials Management, Tucson, Arizona, July 2009.
- [7] Young S. Ham et. al, "Experimental Validation of the Methodology for Partial Defect Verification of the Pressurized Water Reactor Spent Fuel Assemblies, 50th Annual Meeting of the Institute of Nuclear Materials Management, Tucson, Arizona, July 2009.
- [8] Doug Reilly et. al., Passive Nondestructive Assay of Nuclear Materials, National Technical Services, 1991
- [9] ORIGEN-ARP, Versions 5.0/5.1.01, Isotope Generation and Depletion Code, CCC-725/732, Radiation Safety Information Computational Center. May 2004/March 2007.
- [10] X5-Monte Carlo Team, MCNP-A general Monte Carlo N-Particle Transport Code, Version 5.1.40, Los Alamos National Laboratory, February 2006.